Carbon Composite Bipolar Plates

T. M. Besmann (Primary Contact), J.W. Klett, and J. J. Henry, Jr.
Surface Processing and Mechanics Group and Carbon and Insulating Materials Group
Oak Ridge National Laboratory, P.O. Box 2008, MS 6063, Bldg. 4515
Oak Ridge, TN 37831-6063
Phone: (865) 574-6852; Fax: (865) 574-6918; E-mail: besmanntm@ornl.gov

DOE Technology Development Manager: Nancy L. Garland
Phone: (202) 586-5673; Fax: (202) 586-9811; E-mail: Nancy.Garland@ee.doe.gov

ORNL Technical Advisor: David Stinton
Phone: (865) 574-4556; Fax: (865) 574-6918; E-mail: stintondp@ornl.gov

Objectives

- Develop a slurry molded carbon fiber material with a carbon chemical vapor infiltrated sealed surface as a bipolar plate.
- Collaborate with manufacturer/licensee with regard to testing and manufacturing of such components.

Technical Barriers

This project addresses the following technical barrier from the Fuel Cells section of the Hydrogen, Fuel Cells and Infrastructure Technologies Program Multi-Year R,D&D Plan

- O. Stack Material and Manufacturing Cost

Approach

- Fabricate fibrous component pre-forms for the bipolar plate by slurry molding techniques using carbon fibers of appropriate lengths.
- Fabricate hermetic plates using a final seal with chemical vapor infiltrated carbon.
- Develop commercial-scale components for evaluation.

Accomplishments

- In coordination with licensee, have reduced thickness of plates from 2.5 mm to 1.5 mm. Determined influence of chemical vapor infiltration (CVI) temperature on depth of infiltration.
- Further characterized and measured mechanical properties of carbon composite plate material.
- Determined electronic properties, including effect of surface roughness on resistance.
- Developed initial model of chemical vapor infiltration process that is scalable to production. The model was utilized to improve the flow pattern of gas used to deposit carbon on the plate.

Future Directions

- Support continued scale-up of the licensee's operation through modeling of fabrication processes.
- Optimize mechanical properties through combination of pre-form material and CVI conditions.

Introduction

The challenges for PEMFC technology for automobiles lie in reducing the cost and weight of the fuel stack, an impediment to which is the cost and weight of the bipolar plate. The bipolar plate is the electrode plate that separates individual cells in a stack. A stack is formed when multiple cells are aligned one after another so as to work in series, with the bipolar plate providing an electrode for the cells on either side. The reference material for the bipolar plate is high-density graphite with machined flow channels. Both material and machining costs for graphite, however, are prohibitive for many fuel cell applications, and this has led to substantial development efforts to replace graphite. The requirements for a bipolar plate are stringent, including low-cost materials and processing, light weight, thin (<3mm), sufficient mechanical integrity, high surface and bulk electronic conductivity, low permeability (boundary between fuel and oxidant), and high corrosion resistance (in the moist atmosphere of the cell).

The bipolar plate approach developed at Oak Ridge National Laboratory (ORNL) uses a low-cost, slurry-molding process to produce a carbon-fiber pre-form. The molded, carbon-fiber component could have an inherent volume for diffusing fuel or air to the electrolyte surface or impressed flow-field channels. The bipolar plate is made hermetic through chemical vapor infiltration (CVI) with carbon. The infiltrated carbon also serves to make the component highly conductive.

The work during FY 2003 included efforts to reduce the thickness of the bipolar plate. Other research focused on determining the properties of bipolar plates prepared from pre-form material supplied by Porvair Fuel Cell Technology (the licensee) as well as ORNL, as a function of processing. This includes strength and fracture toughness, the depth of penetration of infiltrated carbon, and the effect of surface roughness on conductivity/resistivity. Also developed during this period was a first-order model of the CVI process based on the ORNL CVI system. These efforts were in cooperation with and in support of the work at Porvair to scale up and commercialize the technology.

Approach

At Porvair Fuel Cell Technology, pre-forms were produced that were reduced from 2.5 mm in thickness to 1.5 mm through an increased concentration of particulate material in the pre-form that resulted in a higher green density (density before infiltration with carbon). The effect of this higher green density on infiltration was investigated through intercomparisons with ORNL material, which did not contain particulates and was of lower density. Void volume as a function of through-thickness position was determined by digital analysis of images of polished cross-sections. The effect of processing conditions (CVI temperature) on density gradients through the plates were similarly investigated.

The mechanical properties of the plates were determined through notch sensitivity. Holes of differing sizes were machined in plate specimens and tested under tension to failure. The characterization of the variation of strength with hole size provides information on fracture toughness. Ultimate strength is found from the zero intercept of the strength versus the ratio of hole diameter to sample width.

The electrical properties as a function of surface roughness may be an issue in fuel cell stacks. This was investigated by producing samples with differing surface roughnesses due to different CVI temperatures, and by also evaluating polished and uninfiltrated samples. Two-point and four-point measurements were obtained which provide contact resistance and bulk conductivity, respectively.

In order to allow efficient scale-up of the CVI process, a computational fluid dynamics model of the process, coupled with chemical kinetic information, was developed. This used the existing CVI system geometry and conditions at ORNL.

Results

Porvair has successfully reduced the thickness of the bipolar plates through a high particulate content giving high green strength (strength before infiltration with carbon). Determination of the through-thickness density gradient of the infiltrated Porvair material reveals that little carbon is deposited within the material and that the overcoating and sealing of the surface happens early during infiltration (Figure 1). This is in contrast to the original ORNL pre-form material that consists entirely of fiber and carbonized phenolic, and has more than twice the uninfiltrated void volume. Thus, after infiltration, especially at higher temperatures where the ORNL material is overcoated and sealed relatively rapidly, the ORNL component has a large density gradient, whereas the infiltrated Porvair pre-form has a high-density outer coating and fairly uniform internal density. Lower temperature and longer infiltration times do not modify this behavior for the Porvair material, but do result in higher density, more uniform ORNL plates. All infiltrated plates are hermetic.

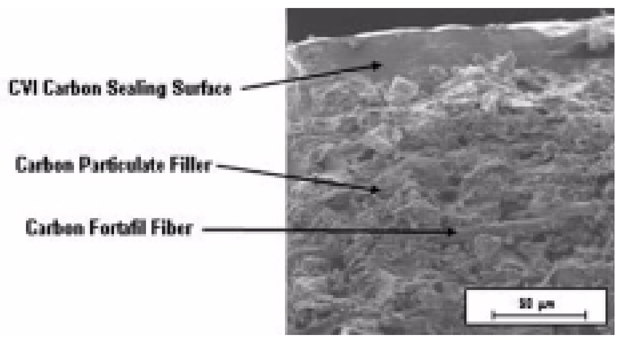

Figure 1. Fracture Surface of a Sealed Bipolar Plate Produced with Porvair Pre-Form Material Showing the Particulate Filled Interior and the CVI Carbon Coating on the Surface

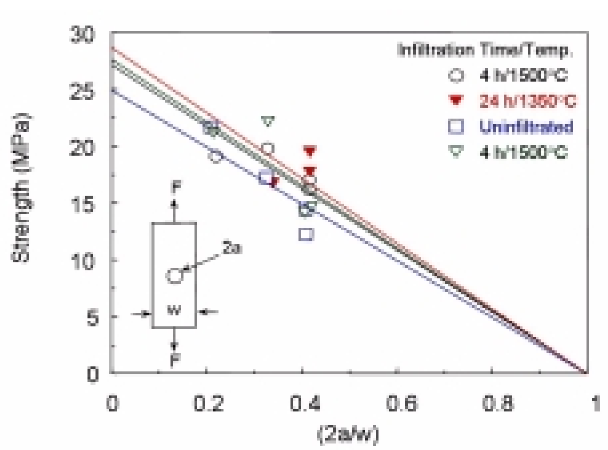

Figure 2. Plot of Measured Strength Versus the Ratio of Hole Size to Sample Width Illustrating Ultimate Strength and Notch-Insenstivity

The strength and fracture toughness of the bipolar plate material were evaluated using the variable hole tensile test. The results of the tensile curves (Figure 2) reveal that the material is relatively notch insensitive. The strengths indicated by the zero intercepts (Figure 2) range from 25 to 28 MPa.

Measurements of contact resistance show a weak dependence on surface roughness. Table 1 shows the results where only the polished samples indicate a measurably lower contact resistance, with the exception of the uninfiltrated material which had fewer interfaces that could contribute to ohmic losses. High bulk conductivity is also indicated.

The computational fluid dynamics modeling revealed flow patterns within the CVI reactor. This included differences in the flow above and below the bipolar plate. This information was used to modify the inlet geometry so that the flow was significantly more uniform, and therefore deposition rates on the top and bottom of the plates were more uniform. Figure 3 illustrates the effect of improving the flow geometry.

Conclusions

- Increasing the particulate content of the pre-form allows a reduction in bipolar plate thickness, more uniform density, and easier/more rapid sealing via CVI.
- Mechanical property measurements indicate the bipolar plate material is notch-insensitive, and therefore relatively tough, and has good strength.
- Electrical measurements indicate low contact resistance and high bulk conductivity, with a weak dependence on surface roughness.

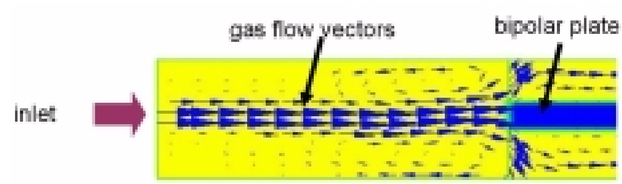

Figure 3. Computed Flow Pattern of Reactive Gases Around a Bipolar Plate in the ORNL CVI Reactor

Table 1. Two- and Four-Point Probe Electrical Measurements

Infiltration Treatment	Sourface Roughness Rz µm	Resistance 2 Probe DC ohms	Conductivity 4 Probe V/I σ S/cm
4 hours at 1500°C	8.83	1.86	365
24 hours at 1350°C	12.9	1.37	417
Uninfiltrated	13.5	0.86	369
4 hours at 1500°C	10.2	2.16	386
24 hours at 1350°C	14.3	1.57	383
24 hours at 1350°C	2.9	0.84	408

- Computational fluid dynamics applied to the CVI process at ORNL indicated means for increasing plate infiltration uniformity.

FY 2003 Publications/Presentations

1. T. M. Besmann, J. J. Henry, Jr., J. Klett, D. Haack, and K. Butcher, "Optimization of a Carbon Composite Bipolar Plate For PEM Fuel Cells," Proc. Materials Research Society, Vol. 756

Special Recognitions & Awards/Patents Issued

1. Year 2002 National Federal Laboratory Consortium "Excellence in Technology Transfer Award."

2. Year 2002 Southeast Region of the Federal Laboratory Consortium "Excellence in Technology Transfer Award."